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THEORETICAL STUDIES OF SOLAR-PUMPED LASERS

By

Wynford L. Harries, Principal Investigator

Progress Report  
For the period January 16, 1983 to July 15, 1983

Prepared for the  
National Aeronautics and Space Administration  
Langley Research Center  
Hampton, Virginia 23665

Under  
Research Grant NSG 1568  
John Wilson, Technical Monitor  
Space Systems Division

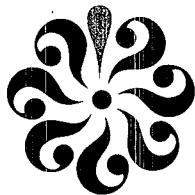


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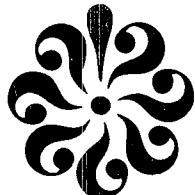
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# THEORETICAL STUDIES OF SOLAR-PUMPED LASERS

By

Wynford L. Harries\*

## SUMMARY

The work performed during the period January 1983 to July 1983 can be divided into three parts. First the theoretical investigations of non-dissociative type lasers was continued and a preliminary study was made by Na<sub>2</sub> and Li<sub>2</sub> metal vapor lasers, and their efficiencies estimated. This part of the work is included under "Metallic Vapor Lasers as Solar Energy Converters".

Whilst this work was in progress it was realized that the Air Force was interested in solar pumped lasers emitting in the blue-green (460-530 nm) for communications and surveillance. Dissociative lasers such as Br<sub>2</sub> - CO<sub>2</sub>-He(1), IBr(2) or n-C<sub>3</sub>F<sub>7</sub>I(3) are limited in their wavelength range by the energy of the excited atom Br\* or I\*, and emit in the infrared. Non-dissociative lasers already under study in principle can emit in the blue-green. Accordingly a "white paper" was written for the Air Force. The purpose of the white paper was to explain the mechanism of non-dissociative lasers and outline experimental measurements of fundamental constants. If the Air Force is interested then funding will be sought for a program initially to be placed in Langley. The first part of the white paper was a condensed version of "Metallic Vapor Lasers as Solar Energy Converters" and is not included here. The latter part was a compendium of experimental measurements and is included in the section "Proposed Measurements of Rate Coefficients and Cross Sections for Feasibility Studies of Solar Pumped Lasers."

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The third part of the work was an investigation of the output power to weight ratio, a figure of merit for solar pumped lasers in general. The results are shown in the section "The Ratio of the Output Power to Weight for Solar Pumped Lasers."

The paper "Kinetic Modeling of an IBr Solar Pumped Laser" by W. L. Harries and W. E. Meador has been accepted in Space Solar Power Review.

## METALLIC VAPOR LASERS AS SOLAR ENERGY CONVERTERS

### ABSTRACT

Metallic vapor lasers of  $\text{Na}_2$  and  $\text{Li}_2$  are examined as solar energy converters. The absorbed photons cause transitions to vibrational-rotational levels in an upper electronic state. With broad band absorption the resultant levels can have quantum numbers considerably higher than the upper lasing level. The excited molecule then relaxes to the upper lasing level which is one of the lower vibrational levels in the upper electronic state. The relaxation occurs from collisions, provided the molecule is not quenched into the ground level electronic state. Lasing occurs with a transition to a vibrational level in the lower electronic state. Rough estimates of solar power efficiencies are 1 percent for  $\text{Na}_2$  and probably a similar figure for  $\text{Li}_2$ . The non-dissociative lasers form a family distinct from materials which dissociate to yield an excited atom. The high working temperatures enable the area of the heat radiator to be small, thus permitting a favorable output power to weight ratio. In contrast to the dissociative lasers which emit in the infrared, the non-dissociative lasers can emit in the visible.

### INTRODUCTION

The possibility of solar energy conversion using gas lasers has been recently considered (1-4). Two classes of laser were studied theoretically: first where the absorber and lasant were different materials, e.g., a  $\text{Br}_2$  -  $\text{CO}_2$  - He laser (the  $\text{Br}_2$  absorbed the photons and handed over the energy to  $\text{CO}_2$ , which lased) and second, where only one material was used, e.g., an  $\text{IBr}$  laser (2, 3). However, the theoretical solar power efficiencies were low - 0.5 and 1.2 percent respectively. In addition, lasing has been accomplished experimentally by pumping  $\text{n-C}_3\text{F}_7\text{I}$  with a solar simulator, but the efficiency was again low, about 0.1 percent (4).

In the above cases, the absorption of photons resulted in photodissociation with the production of excited atoms  $\text{Br}^*$  or  $\text{I}^*$ . Whereas relatively

few diatomic molecules can be dissociated by light near the solar peak, there are a great many that can absorb a photon and be raised to one of the vibrational-rotational levels of an upper electronic state without dissociation. Lasing could then occur as a transition to one of the vibrational levels of the ground electronic state.

Recently, Wellegehausen (5,6) listed a number of optically pumped dimer lasers including the metal vapors  $\text{Li}_2$ ,  $\text{Na}_2$ ,  $\text{Bi}_2$ , and  $\text{Te}_2$  which lased without dissociation. Several hundred lines were reported in the range 400-1350 nm with emission bandwidths of less than 3 GHz. The vapors were pumped with frequency doubled Neodymium lasers at 532.5 nm and with Argon lasers at 472.7 nm. In view of the low thresholds and high measured efficiencies and because absorption occurred near the peak of the solar spectrum, the possibility of using these vapors as solar energy converters arises. The quoted efficiencies were up to 15 percent, but for solar energy conversion the overall efficiency should include the solar efficiency (fraction of the solar radiance used) which is usually below 20 percent (2). Vaporizing the metals would be achieved by solar concentrators and the lasers would run at temperatures of around 1000 K. The high temperatures would also reduce the area requirement of the heat dissipation surface an important factor in the output power to weight ratio, and an advantage over the  $\text{IBr}$  laser.

It is the purpose of this paper to make a preliminary estimate of the operation of metal vapor non-dissociative solar pumped lasers, especially  $\text{Na}_2$  and  $\text{Li}_2$  which experimentally have the lowest threshold powers. These lasers form a family distinct from dissociative lasers. The question of efficiency is of particular importance. The case of  $\text{Na}_2$  will be studied first because the reaction rate constants are more completely available.

#### MECHANISM OF THE LASER

The mode of lasing is illustrated in the energy diagram of a dimer (figure 1). Photon absorption results in transitions from vibrational levels in the lower electronic state to those in the upper, according to the Franck-Condon principle. At high temperatures, vibrational levels above the ground state of the lower electronic level will be occupied resulting in increased absorption bandwidth.

The difference between the operation of a solar pumped laser (which requires broad band absorption) and the laser pumped experiments is that in the former, transitions can occur to levels with much higher vibrational quantum numbers  $v'$  in the upper electronic state than the upper lasing level of quantum number  $v_u'$ , which is usually 0, 1 or 2. Not all of the excited molecules relax to level  $v_u'$  - a disadvantage, as the absorbed energy is wasted. A molecule raised to level  $v'$  can either descend to level  $v_u'$  by collisions (or for heteronuclear molecules by radiating) or leave the upper electronic level by quenching. (The quenching mechanism is taken here as any collisional process which causes the molecule to leave the upper electronic state without the emission of a photon.) The probability of arriving at the upper laser level turns out to be a critical feature in the laser efficiency.

### The $\text{Na}_2$ METALLIC VAPOR LASER

#### Photon Absorption

The Einstein coefficient for transitions from the lower level 1 to the upper level 3 (figure 1) of the  $\text{Na}_2$  laser is  $A_{13} = 10^7 \text{ s}^{-1}$  (5). The absorption cross section is large,  $\sigma_a = A_{13}/8\pi c v_{13}^2 = 3 \times 10^{-14} \text{ cm}^2$  for absorption around 500 nm. The absorption length  $L$  then turns out to be  $10^{-3}/p$  (cm) where  $p$  is the pressure of  $\text{Na}_2$  vapor in torr. For reasonable active lengths, low pressures are necessary. The estimate of  $L$  is consistent with lasing in supersonic beams of  $\text{Na}_2$  with an active length of order 1 mm. However, the fraction of the vapor which is in the form of  $\text{Na}_2$  is about 0.04 by number, with the remainder in the form of Na at around 700K where the laser operated (7).

#### Choice of the Lasing Levels

The lower laser level 2 (figure 1) must have a population of less than half that of the ground level 1 so that it can be adequately depleted (7). The reason is that the maximum population density achievable in level 3 with a strong pump source is  $\bar{n}_3 = \bar{n}_1$ . This means that (neglecting statistical weights) the density of level 1 at temperature  $T$  is divided into 2 between

levels 3 and 1:  $\bar{n}_3 = \bar{n}_1 = n_1(T)/2$  where  $n_1(T)$  is the normal Boltzmann population. Population inversion is therefore possible only if  $n_3 = n_1(T)/2 > n_2(T)$  (5). At 800 K the lowest value of the quantum number  $v''$  for level 2 is  $v'' = 3$ . Lasing to the  $v'' = 3$  level would originate near the  $v' = 0$  level in the upper electronic state. The low value of  $v'$  is helpful in utilizing most of the populations of states with higher  $v'$ , provided they can relax downward. Level 2 must be depleted by collisions thus requiring a buffer gas such as Argon.

The closeness of the vibrational energy levels in both ground and electronically excited states raises the question of whether there could be lasing from more than one upper level to a corresponding lower level. The two transitions would have to be equal in frequency within the emission bandwidth of 3 GHz but this is unlikely because the spacing of the vibrational levels is around  $6 \times 10^{12}$  Hz or 2000 times greater.

#### Requirement for Collisional Relaxation in the Upper Electronic State

The collisional relaxation rate for levels near the upper laser level must exceed the spontaneous emission rate (neglecting quenching for the moment), otherwise the density of molecules in the upper level will not increase before threshold. If Argon collisions dominate, the requirement is:

$$(Na_2)(Ar)\sigma_3 v_3 > (Na_2)\gamma_3^0 \quad (1)$$

where the quantities in parenthesis are densities,  $\sigma_3$  the cross section,  $v_3$  the relative velocity and  $\gamma_3^0$  the spontaneous emission rate. With  $\sigma_3 = 5 \times 10^{-15}$  cm<sup>2</sup>,  $v_3 = 7 \times 10^4$  cms<sup>-1</sup> and  $\gamma_3^0 = 1.4 \times 10^8$  s<sup>-1</sup> (3) the inequality is satisfied for Argon pressures greater than 11 torr.

#### Requirement for Stimulated Emission to Exceed Collisional Relaxation

Lasing requires the stimulated emission rate to exceed the collisional relaxation rate out of  $v'_u$  of the upper state. The stimulated emission depends on the inverted population density and emission cross section. Assuming the upper level to be  $B^1\Pi_u$ , with an angular momentum quantum

number of 1, and the lower level to be  $X^1\Sigma_g^+$  with no angular momentum, then the multiplicities for the upper and lower levels are  $g_u = 2$  and  $g_e = 1$ . The inverted populated density is:

$$\Delta n = (Na_2, v'_u) - 2(Na_2, v''_l) \quad (2)$$

where the quantities in parentheses are the densities in the upper and lower laser levels respectively. It follows that  $(Na_2, v'_u)$  must exceed  $2(Na_2, v''_l)$  for lasing. The emission cross section  $\sigma_e = \lambda_e^4 A_{32} / 4\pi^2 c \Delta\lambda_e \text{ cm}^2$  is evaluated at  $1.3 \times 10^{-12} \text{ cm}^2$ . Here  $\lambda_e$  is the emission wavelength 525 nm,  $A_{32} = 1.9 \times 10^7 \text{ s}^{-1}$  and the emission bandwidth  $\Delta\lambda_e = 10 \text{ nm}$  (corresponding to Doppler broadening at 800 K). The value agrees roughly with the quoted value  $3 \times 10^{-12} \text{ cm}^2$  (5). The requirement that stimulated emission exceeds depletion by collisions of the upper level is:

$$nc \sigma_e \Delta n > (Na_2, v'_u) (Ar) \sigma_3 v_3 \quad (3)$$

where  $n$  is the photon density during lasing. For pressures corresponding to 0.04 torr of  $Na_2$  and 0.96 torr of Na and 60 torr of Argon, this is equivalent to requiring  $n\Delta n / (Na_2, v'_u) > 2 \times 10^{10} (\text{cm}^{-3})$  which should be easily satisfied if  $n\Delta n > 10^{12}$  and less than 10 percent of the  $Na_2$  atoms were in the upper laser level.

#### Requirement for Collisional Relaxation in the Lower Laser Level

For CW operation the total relaxation rate for the lower laser level must be greater than the spontaneous transition rate for a specific transition  $3 \rightarrow 2$  or  $(Na_2, v''_l) (Ar) \sigma_3 v_3 > (Na_2, v'_u) A_{32}$ . With the above numbers and  $A_{32} = 1.9 \times 10^7 \text{ s}^{-1}$ , the condition is equal to  $(Na_2, v'_u) / (Na_2, v''_l) < 40$ , which is easily realized.

#### Probability of Filling the Upper Laser Level

The energy level diagram of  $Na_2$  (figure 2) (8) shows numerous vibrational levels (of quantum numbers  $v'$  and  $v''$  in the upper and lower electronic states) spaced closely together. At 800 K, there is a one percent probability of filling the  $v'' = 10$  vibrational state, and an absorption transition then populates an upper vibrational level with  $v'$  about 30. The energy must be transferred to the upper laser level without being dissipated

from the molecule. A rough estimate of the probability of arriving at the upper lasing level  $v'_u$  from a higher vibrational level  $v'$  will now be made.

The molecule can drop from a higher to a lower vibrational level by collisions. A model by Landau and Teller (9) enables an estimate to be made of the number of collisions required for a transfer of vibrational into translational energy and the number is of order 1 for  $\text{Na}_2$  (App. I). The vibrational levels in the upper electronic state are about  $120 \text{ cm}^{-1}$  apart corresponding to 170 K. If the gas temperature is 800 K, assuming the energy exchanged is equal to half the kinetic energy, then the jump in vibrational levels corresponds to  $\Delta v' = \pm 2$ , and the transition can be either upwards or downwards. Such collisions can be due either to  $\text{Na}_2$  molecules or Argon atoms ( $\sigma_3 = 5 \times 10^{-15}$ ) (5). Collisions of this kind leave the molecule still in a vibrational level in the upper electronic state.

The rate equation for relaxing from level  $v'$  to level  $(v' \pm \Delta v')$  is:

$$\frac{d}{dt_r} (\text{Na}_2, v') = (\text{Na}_2, v') (\text{Na}_2) \sigma_1 v_1 + (\text{Na}_2, v') (\text{Ar}) \sigma_3 v_3 \quad (4)$$

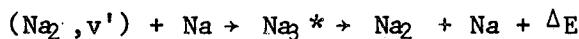
where  $(\text{Na}_2, v')$  is the density of  $\text{Na}_2$  molecules of vibrational quantum level  $v'$ , and  $\sigma_1 = 10^{-4} \text{ cm}^2$  and  $\sigma_3 = 5 \times 10^{-15} \text{ cm}^2$  are the cross sections for collisions with  $\text{Na}_2$  molecules of relative speed  $v_1$  and Argon of relative speed  $v_3$ . All collisions with Na are assumed to result in quenching (5), (see below).

The processes which cause the molecule to relax from the upper to the lower electronic state (apart from stimulated emission) are spontaneous emission of a photon (rate coefficient  $\gamma = 1.4 \times 10^8 \text{ s}^{-1}$ ) (5) and quenching. The spontaneous emission rate is  $\gamma^0 = 1.4 \times 10^8 \text{ s}^{-1}$  (5). Hence, the rate of spontaneous emission from the upper electronic state to the ground state is:

$$\frac{d}{dt_s} (\text{Na}_2, v') = (\text{Na}_2, v') \gamma_3^0 \quad (5)$$

Quenching can be due to  $\text{Na}_2$  or Na. The quenching cross section for  $\text{Na}_2$   $\sigma_4$  is probably around  $10^{-14} \text{ cm}^2$ , but the fraction of  $\text{Na}_2$  is small. The

quenching cross section of Argon is assumed to be negligible as it has no degrees of freedom. The cross section for Na is large ( $\sigma_5 = 6 \times 10^{-14} \text{ cm}^2$ ) (5). The reason is that the Na forms complexes with  $\text{Na}_2$  which have internal degrees of freedom and can absorb the energy drop of  $2 \times 10^4 \text{ cm}^{-1}$ , before splitting up into an  $\text{Na}_2$  molecule in the lower electronic level possessing vibrational and rotational energy and an Na atom, both with high kinetic energy.



The quenching rate from vibrational quantum level  $v'$  is:

$$\frac{d}{dt}_Q (\text{Na}_2, v') = (\text{Na}_2, v') (\text{Na}_2) \sigma_4 v_1 + (\text{Na}_2, v') (\text{Na}) \sigma_5 v_2 \quad (6)$$

and  $v_2$  is the relative speed of Na.

The probability that a collision will leave the molecule in the upper electronic state is then:

$$f = \frac{d}{dt}_r (\text{Na}_2, v') / \left\{ \frac{d}{dt}_r (\text{Na}_2, v') + \frac{d}{dt}_s (\text{Na}_2, v') + \frac{d}{dt}_Q (\text{Na}_2, v') \right\} \quad (7)$$

The  $\text{Na}_2$  laser experiments were performed at a total pressure of Na +  $\text{Na}_2$  of about one torr with Argon pressures of many tens of torr. A plot of  $f$  vs Argon pressure for these conditions for different percentages by number of  $\text{Na}_2$  (from 2 to 20 percent) (figure 3) shows that  $f$  increases with Argon pressure but is not very sensitive to the  $\text{Na}_2$  fraction (and therefore also to  $\sigma_1$  and  $\sigma_4$ ).

The experiments with laser pumped  $\text{Na}_2$  vapor showed that the power output dropped to zero at about 70 torr of Argon, and the reason quoted was the increase of the relaxation rate of the upper laser level due to Argon collisions (5). It is possible this effect would not occur in a solar pumped laser, because the relaxation process would supply the upper laser level from levels above  $v_u'$ , as well as deplete it. Should pressures above 70 torr be unworkable then  $f \leq 0.7$ . (figure 2). On the other hand, if it were possible to use 300 torr of Argon then  $f \approx 0.9$ . The quenching by Argon is assumed small (7).

There are 88 vibrational levels in the  $B^1\pi_u$  state, and levels up to about  $n = 30$  will be occupied. The probability of arriving at  $v'_u = 0$  from a higher  $v'$  is next roughly estimated. The spacing of the vibrational levels is assumed constant at  $\omega_e \text{ cm}^{-1}$ . The collisions that transfer vibrational to translational energy can raise or lower the molecule from vibrational level  $v'$  to  $v' \pm \Delta v'$ . The molecule, as it changes its vibrational levels through collisions, performs a one dimensional random walk in energy space with a step length  $\Delta v' \omega_e (\text{cm}^{-1})$  and has to travel a distance  $(v' - v'_u) \omega_e$  to arrive at the upper lasing level  $v'_u$ . The "diffusion coefficient" is  $(\Delta v' \omega_e)^2 v_c$  where  $v_c$  is the collision frequency. The diffusion equation  $(v' - v'_u)^2 \omega_e^2 = (\Delta n \omega_e)^2 v_c t$ , gives the number of collisions  $N = v_c t$  as:

$$N = (v' - v'_u)^2 / (\Delta v')^2 \quad (8)$$

The equation is approximate as it assumes a constant step length, but is sufficient for illustration.

If a fraction  $f$  of the molecules stays in the upper electronic state of each collision, then in  $N$  collisions the fraction remaining is on the average:

$$F = f^N \quad (9)$$

e.g., if for  $\text{Na}_2$   $f = 0.7$ ,  $\Delta v' = 2$ ,  $v' - v'_u = 10$ , then  $F = 1.3 \times 10^{-4}$ . But if  $f = 0.9$  then  $F = 0.39$ .

Assuming all the vibrational levels above  $v'_u$  are filled with equal probability (not too inaccurate an assumption because only levels near  $v'_u$  contribute) then the contribution to the level  $v'_u$  from all the levels is

$$S = \sum_{v'=v'_u+1}^{\infty} f^{[(v'-v'_u)/\Delta v']^2} \quad (10)$$

The summation converges rapidly with increasing  $v'$  especially if  $f$  is

small. For example, if  $f = 0.7$ ,  $\Delta v' = 2$  then  $S = 2.47$ , but if  $f = 0.9$  then  $S = 4.96$ . This is equivalent to saying that the first 2, and 5 levels respectively are the only ones that contribute to the upper lasing level.

If levels above  $n = 5$  are essentially unusable for lasing, then figure 2 shows the absorption bandwidth would correspond to 15 nm, the region between 476.6 to 490.6 nm. The solar efficiency  $\eta_s$  for this narrow region is less than two percent.

#### Overall Efficiency of $\text{Na}_2$ Vapor Laser

The absorption efficiency  $\eta_A$  can be made to approach unity with very low  $\text{Na}_2$  pressure if a path length of 1 cm is used. A rough estimate of the fraction of molecules of vibrational levels  $v' \leq 5$  that arrive at the  $v_u' = 0$  level can be made using equation 10 which gives a value of about 0.5 if  $f = 0.7$ . We may call this a kinetic efficiency  $\eta_K$ . The quantum efficiency  $\eta_Q$  here approaches unity. Thus the overall efficiency  $\eta = \eta_s \eta_A \eta_K \eta_Q$  is about 1 percent. If it be possible to increase the Argon pressure sufficiently to raise  $f$  to 0.9, then the overall solar efficiency would approach 2 percent.

#### THE $\text{Li}_2$ METALLIC VAPOR LASER

The temperature required to operate the  $\text{Li}_2$  laser was reported at 1000 K (5),-easily attainable with large solar collectors. The laser was also reported as having the lowest threshold power. The criteria of sections 3.3, 3.4 and 3.5 should be satisfied for  $\text{Li}_2$  as well as  $\text{Na}_2$ , as the coefficients are probably not very different.

#### Probability of Filling the Upper Laser Level

The critical item in the efficiency calculation is the probability  $F = f^N$  (eq. 9). The energy level diagram for the  $X^1\Sigma_g^+$  and the  $B^1\Pi_u$  states of  $\text{Li}_2$  (8) shows the spacing of the vibrational levels is greater than  $\text{Na}_2$ . At 1000 K there would be one percent of the atoms in the  $v'' = 7$  level and transitions up to about  $v' = 30$  are possible.

The spacing of the vibrational levels in the upper state is about  $250\text{cm}^{-1} = 360\text{ K}$  or approximately half the gas temperature, and hence  $\Delta v'$

here is about 1. Hence  $N = (v' - v'_u)^2$ . The cross section for relaxation from vibrational level  $v'$  by collision is the same order of magnitude for  $\text{Li}_2$  as for  $\text{Na}_2$  (App. I). The rates and cross sections for  $\text{Li}_2$  are not known, but should not be greatly different from  $\text{Na}_2$ .

#### Efficiency of the $\text{Li}_2$ Vapor Laser

At 1000 K vibrational level  $v'' = 7$  will have one percent probability of being occupied and the absorption bandwidth is from 400 to 490 nm. The cross sections and rate coefficients for  $\text{Li}_2$  are not known but it is likely that the overall efficiency would be of the same order as that of  $\text{Na}_2$ .

#### CONCLUSIONS

A class of non-dissociative metallic-vapor dimer lasers is investigated as potential solar energy converters. The high temperatures required for vaporization cause vibrational levels up to about  $v'' = 10$  to be occupied in the lower electronic state. This allows broad band absorption essential for high efficiency, and yields a wide range of levels (up to  $v' = 30$ ) to be occupied in the upper electronic state. The upper laser level is at or near  $v'_u = 0$ , and the probability of the molecule relaxing to  $v'_u$  from higher  $v'$  is small, because it has to do so in a number of collisions, for each of which there is the possibility of being quenched to the lower electronic state. Inclusion of Argon is essential to increase the collisional relaxation rate to overcome quenching. In laser-pumped experiments, the upper limit of Argon pressure in  $\text{Na}_2$  was about 70 torr, in which case the ratio of relaxation to total (inclusion quenching) collisions would be 0.7. This means that for  $\text{Na}_2$  essentially all levels above  $v' = 5$  would contribute little to the lasing power, and that portion of the solar spectrum which caused transitions to  $v' > 5$  was essentially unused. For levels with  $v' < 5$  approximately half the collisions resulted in relaxation to level  $v'_u$ . Even so, the overall solar efficiency was 1 percent a number which compares favorably with 1.2 percent for an IBr solar pumped laser (2). If Argon pressures greater than 70 torr are possible, the efficiency would be increased. The numbers for  $\text{Li}_2$  are not known, but it is probable its efficiency would not be greatly different.

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## APPENDIX I

### Probability of Vibrational to Translational Energy Transfer

The number of collisions  $Z$  required to deexcite a vibrational level  $n$  to a vibrational level  $n \pm \Delta n$  is given by (9)

$$Z = Z_0 Z_{osc} Z_{tr}$$

where  $Z_0$  is a geometrical factor of order 3,  $Z_{osc}$  is dependent on the vibration of the molecule and  $Z_{tr}$  or the translational energy of the impacting particle. Then for a molecule B C Struck by an atom A:

$$Z_{osc} = \frac{2M_B M_C}{M_B^2 + M_C^2} \cdot \frac{M_A + M_B + M_C}{M_A} \frac{1}{2\pi^2} \frac{\theta'}{\theta}$$

where the M's are the respective masses. The quantities  $\theta$  and  $\theta'$  are characteristic temperatures defined by

$$k \theta = h\nu = hc \omega_e$$

where  $k$ ,  $h$ ,  $c$  have their usual meaning and  $\omega_e$  corresponds to the vibrational energy in  $\text{cm}^{-1}$ .

The temperature  $\theta'$  is defined by

$$k \theta' = 16 \pi^4 \tilde{M} \nu^2 L^2$$

where  $\tilde{M}$  is the reduced mass,  $\nu = \omega_e c$  the vibrational frequency, and  $L$  an interaction distance usually taken as 0.2 to 0.3A.

The quantity  $Z_{tr}$  is defined

$$Z_{tr} = \pi^2 \left(\frac{\theta}{\theta'}\right)^2 \sqrt{\frac{3}{2\pi}} \left(\frac{T}{\theta'}\right)^{1/6} \exp \left[ \frac{3}{2} \left(\frac{\theta'}{T}\right)^{1/3} - \frac{\theta}{2T} \right]$$

and  $T$  is the gas temperature in degrees Kelvin.

For the  $B^1\Pi_u$  state of  $\text{Na}_2$  with  $\omega_e = 117.3$ ,  $L = 0.2\text{A}$ , then  $\theta = 168.3\text{K}$ ,  $\theta' = 2.316 \times 10^4 \text{ K}$ .

For  $T = 800\text{K}$ , then  $Z_{osc} = 7$ ,  $Z_{tr} = 3.5 \times 10^{-2}$  which gives  $Z = 0.7$  or on average all collisions with Argon deexcited the  $\text{Na}_2$ .

For the  $B^1\pi_u$  state of  $Li_2$  with  $\omega_e = 255.5$ ,  $L = 0.2\text{A}$  than  $\theta = 367.9\text{K}$  and  $\theta' = 1.552 \times 10^4 \text{K}$ . Again taking Argon and  $T = 1000 \text{ K}$ , then  $Z_{osc} = 2.9$ , and  $Z_{tr} = 8.5 \times 10^{-2}$ . Hence,  $Z \approx 0.7$  and approximately every collision deexcites the  $Li_2$ .

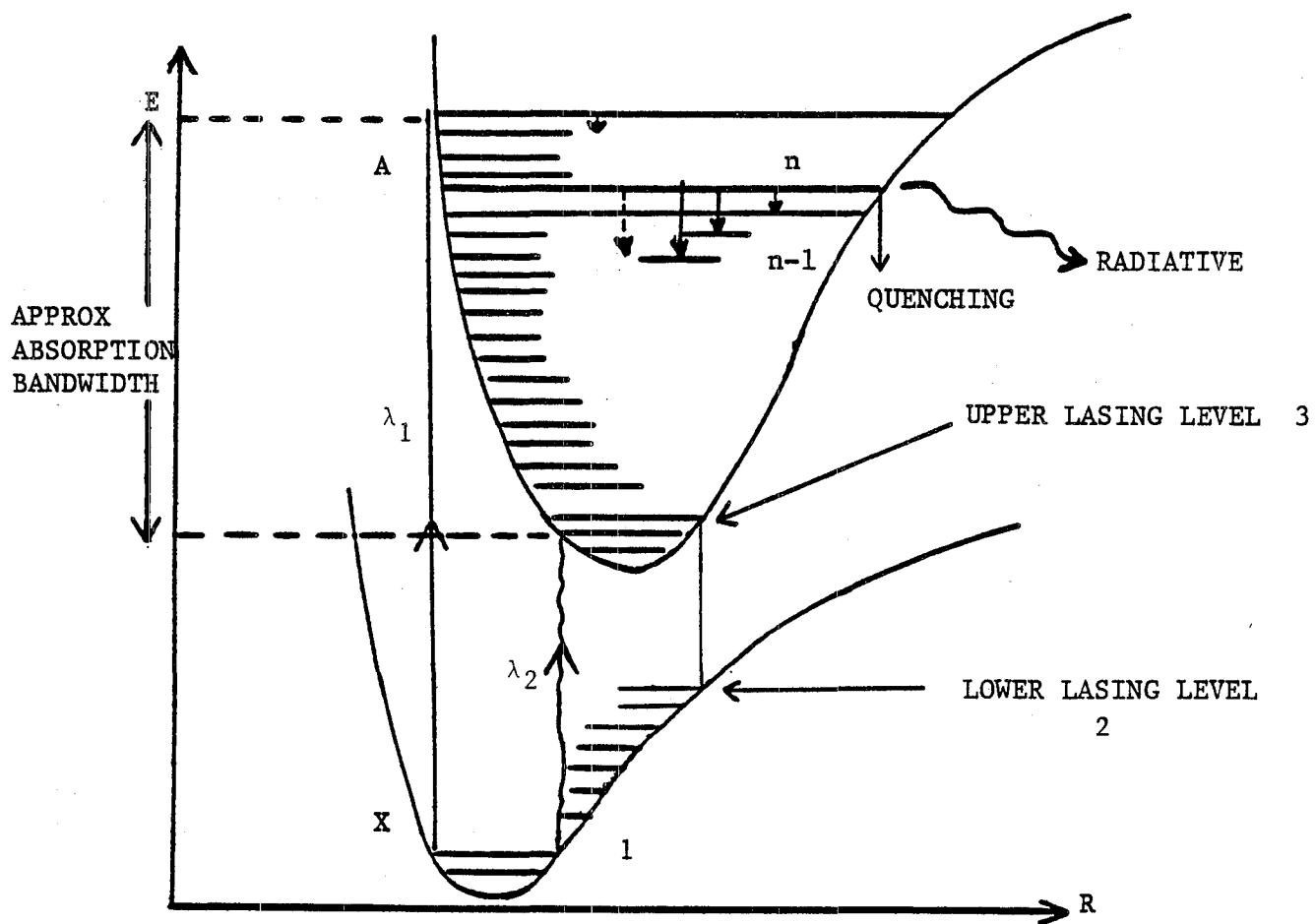


Figure 1. Energy level diagram for a solar pumped dimer molecule  $M_2$ . The vibrational levels only are shown. A molecule raised to level  $v'$  of the upper electronic A state can descend to the  $(v'-1)$ ,  $(v'-2)$ ..., levels by collisions (continuous arrows) (and radiation-dotted arrows for heteronuclear molecules) and remain in the upper electronic state. Otherwise, it may radiate a photon (wave arrow) or be quenched and then drop to the lower electronic X state.

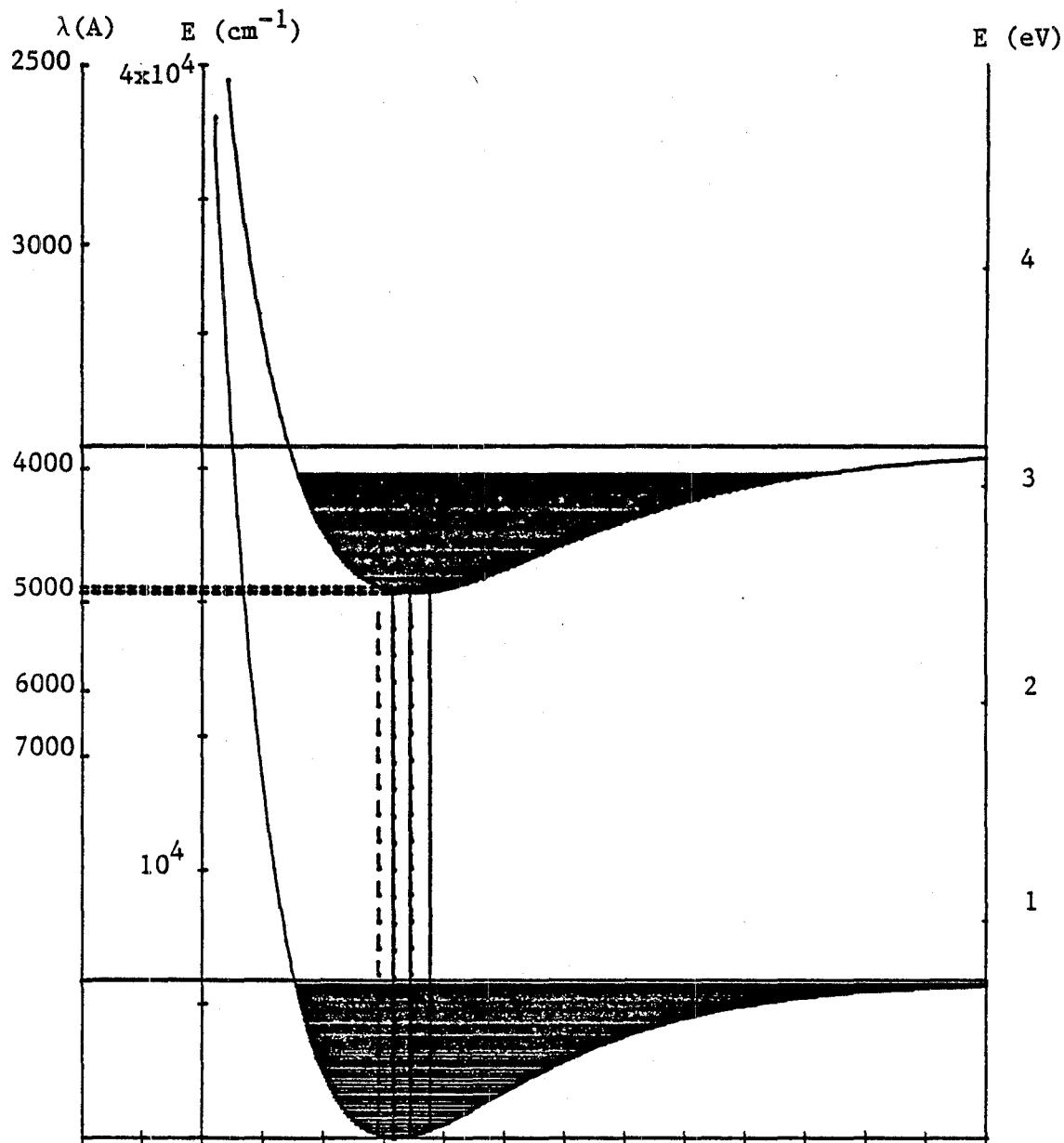


Figure 2. Energy diagram for  $\text{Na}_2$  showing the lower level (X)  $^1\Sigma^+$  and the upper level B  $^1\Pi_u$ . There are 109 vibrational levels in the lower electronic state; only 45 are shown. There are 88 levels in the upper state; 50 are shown. The vertical dotted lines correspond to Franck-Condon transitions upward from the  $v'' = 0$  level. The solid lines correspond to downward transitions from the  $v' = 0$  level in the B  $^1\Pi_u$  state which end in the  $v'' = 3$  level.

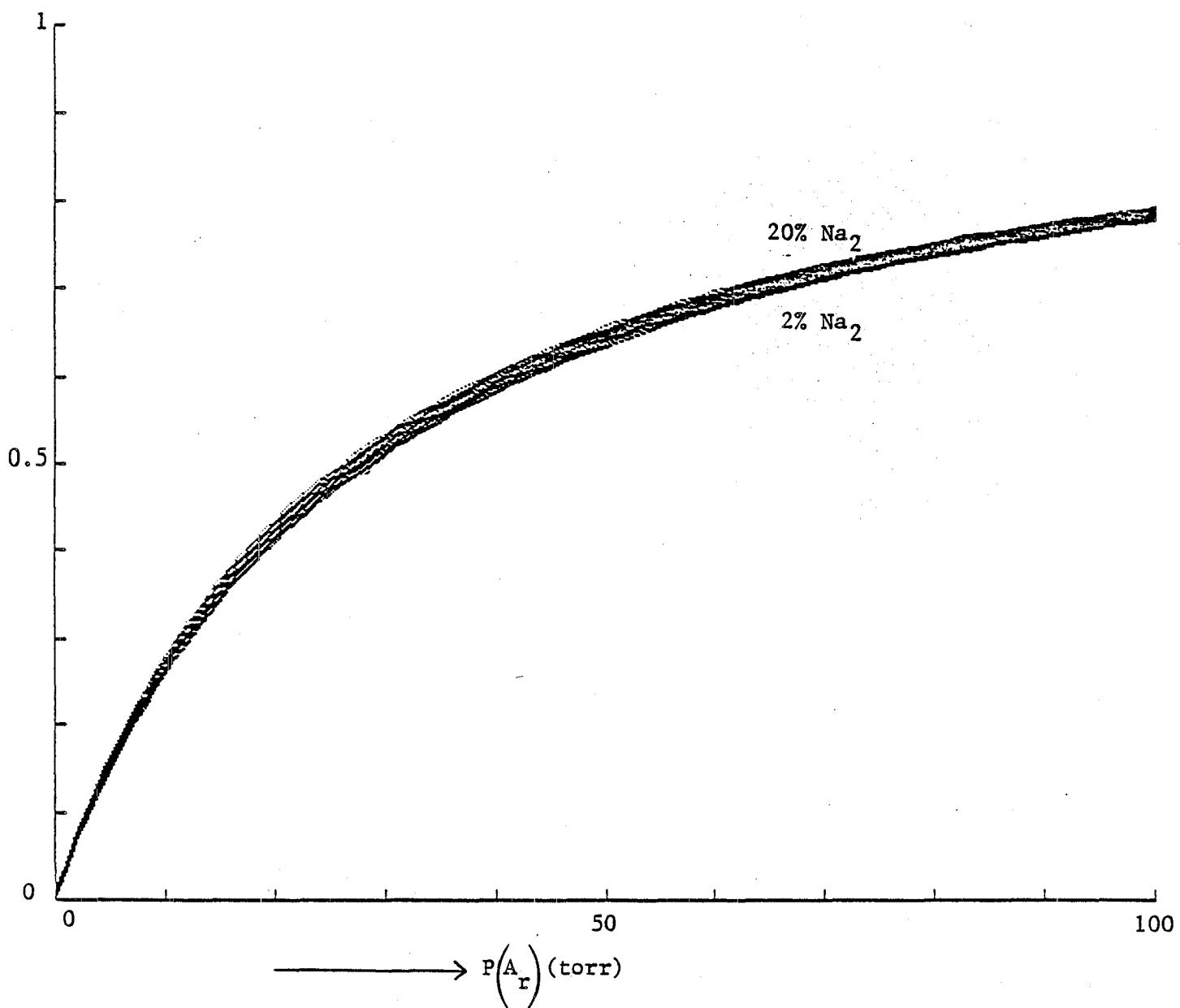


Figure 3. Plot of  $f$ , the fraction of molecules that stay in the upper level per collision vs Argon pressure for different fractions of  $Na_2$  in a total pressure  $Na + Na_2$  of one torr.

PROPOSED MEASUREMENTS OF RATE COEFFICIENTS AND CROSS SECTIONS FOR  
FEASIBILITY STUDIES OF SOLAR PUMPED LASERS

PHYSICAL CONSTANTS REQUIRED FOR LASER MODELING

The case of  $\text{Na}_2$ , with argon as a buffer gas, is taken as an example. The vapor is produced in an oven and consists of Na and  $\text{Na}_2$ . The determination of the ratio  $\text{Na}_2/\text{Na}$ , as a function of temperature, is required to make the measurements effective.

The quantities which involve photon interactions are  $\sigma_a$  the absorption cross section,  $\sigma_e$  the stimulated emission cross section of  $\text{Na}_2$ , the Einstein coefficients  $A_{13}$  from level 1 to level 3, and  $A_{32}$  the lasing transition, and  $\gamma$  the total spontaneous emission coefficient from a level in the upper electronic state. Cross sections involving relaxation of the excited molecule (causing it to relax from a vibrational level  $v'$  in the upper electronic state to  $v' \pm \Delta v'$ ) are  $\sigma'_{r1}$  from collisions with Na,  $\sigma'_{r2}$  from  $\text{Na}_2$ , and  $\sigma'_{rA}$  from argon, and likewise,  $\sigma''_{r1}$ ,  $\sigma''_{r2}$ , and  $\sigma''_{rA}$  in the lower electronic state. The quenching cross sections from collisions with Na,  $\text{Na}_2$ , and argon are  $\sigma_{q1}$ ,  $\sigma_{q2}$ , and  $\sigma_{q3}$ , respectively.

METHODS OF MEASURING THE CONSTANTS

The vapors are formed in an oven and, for a given temperature and total pressure, the ratio of  $\text{Na}_2/\text{Na}$  must be obtained. This can be done by measuring the total pressure  $P_1$  at temperature  $T_1$  and  $P_2$  at temperature  $T_2$ . Simultaneously, the transmitted intensity of a laser beam absorbed by the molecules alone is measured-- $I_1/I_0$ ,  $I_2/I_0$ . The ratio can then be calculated at these two temperatures. The ratio as a function of temperature is known for many dimers (ref. 1). Absorption measurements require that the light beam at no time encounter molecules at a lower temperature than the center of the oven, which totally encloses the heat pipe (fig. 2.1). The absorption measurement yields  $\sigma_a$ , which in turn yields  $A_{13} = 8\pi c \sigma_a v_{13}^2$ , where  $c$  is the velocity of light and  $v_{13}$  is the absorbed frequency. Similary for transition 3 to 2,  $\sigma_{32}$  is measured to give  $A_{32} = A_{23}$ .

Lifetimes of various lines can be estimated from their bandwidth. In addition, the spontaneous emission coefficient,  $\gamma = \tau^{-1}$ , can be obtained by measuring the delay between a pulse of pumping radiation and radiation emitted at right angles (fig. 2.2). This arrangement is similar in principle to that of Ehret and Gerber (ref. 2). The tunable dye laser with a range from 360 to 800 nm emits a 1-ns pulse of 25 kW with a line width of 0.01 nm. The photomultipliers in the polychromator are set to record, simultaneously, the input laser pulse and several emitted lines. The half-meter monochromator gives a resolution of 0.02 nm, adequate to distinguish the various transitions. The photomultiplier circuits are modified for rapid response (the time constants are essentially determined by the capacitance of the last dynode and the resistor in parallel with it). The signals are displayed on a GHz oscilloscope, and the delay (typically  $\approx 10^{-8}$  s) read directly.

The same apparatus can be used to measure the relaxation cross sections  $\sigma_{r1}'$ ,  $\sigma_{r2}'$ , and  $\sigma_{rA}'$ . The molecule is pumped by a nanosecond pulse to, for example,  $v' = 7$ , and emission from  $v' = 6$  to the ground state monitored. The delay in the emission, minus the spontaneous decay time of level 6, is a measure of how many collisions are necessary to lower the vibrational quantum number from 7 to 6. At 120°C, the Na<sub>2</sub>/Na ratio is 0.1 percent, so the effect of Na is calculated first. The ratio is increased up to 10 percent by increasing the temperature to 700°C, and the effect of Na<sub>2</sub> is then estimated. By varying the argon pressure up to 100 times greater than Na + Na<sub>2</sub>, the effect of argon is found.

The relaxation times,  $\sigma_{r1}''$ ,  $\sigma_{r2}''$ , and  $\sigma_{rA}''$  are found in a different manner (fig. 2.3). The beam from the tungsten lamp is sent into the polychromator which is set at the wavelength  $\lambda_2$  for the electronic transition  $v'' - v'$ . The oscilloscope records a steady intensity signal until the laser pulse at  $\lambda_1$  is sent into the vapor. The laser depletes the molecules at level  $v''$  so the transmitted intensity is increased and then, when the pulse is over, it decreases with a decay time corresponding to the level  $v''$  being filled by collisions. Variations of the pressures of Na<sub>2</sub>, Na, and Ar would be performed as before, and  $1/\tau$  plotted vs. P.

The quenching cross sections can be found by pumping the molecules with a 1-ns pulse to level  $v'$  in the upper electronic state and observing the

intensity of the transition  $v' - v''$  vs. time for different pressures and ratios of  $Na_2/Na$  and argon pressures as previously. The leading edge of the pulse gives a direct measurement of filling time of the upper level, the decay time gives the value of  $\tau_Q^{-1}$  for quenching. Plots of  $\tau_Q^{-1}$  vs.  $P$  should be linear.

#### AVAILABILITY OF EQUIPMENT

All equipment, apart from the pulsed dye laser and GHz oscilloscope, is available at NASA/Langley Research Center. The laser and oscilloscope are commercial items. The author has been a grantee at Langley since 1970, and the experiments would be set up there.

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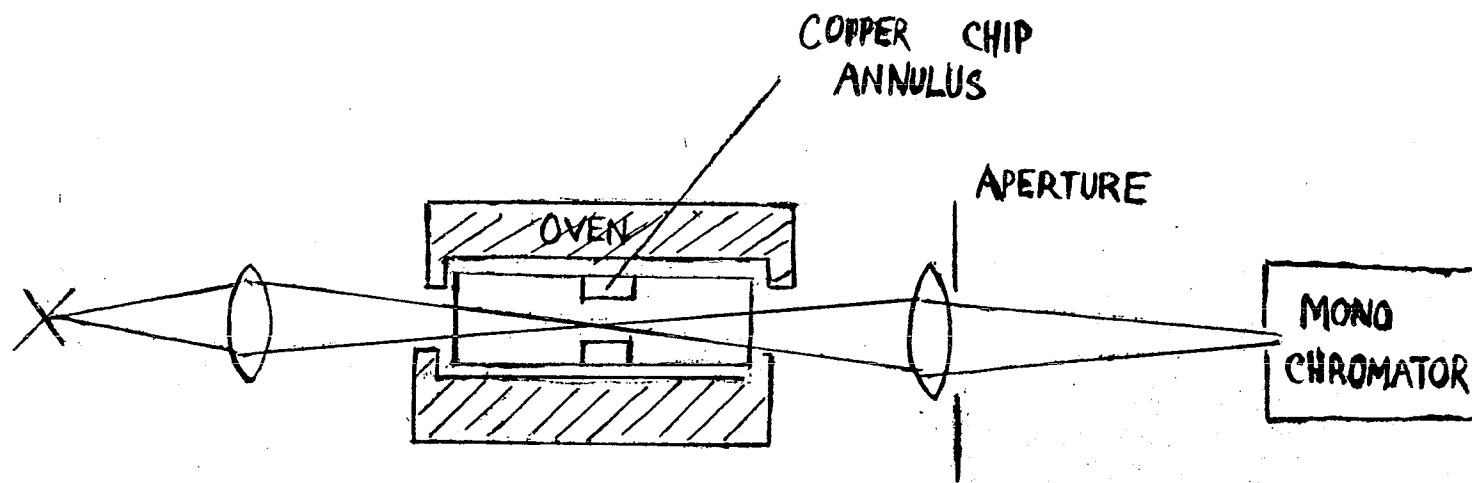


Figure 2.1 Method of measuring absorption (After Lapp and Harris (1))

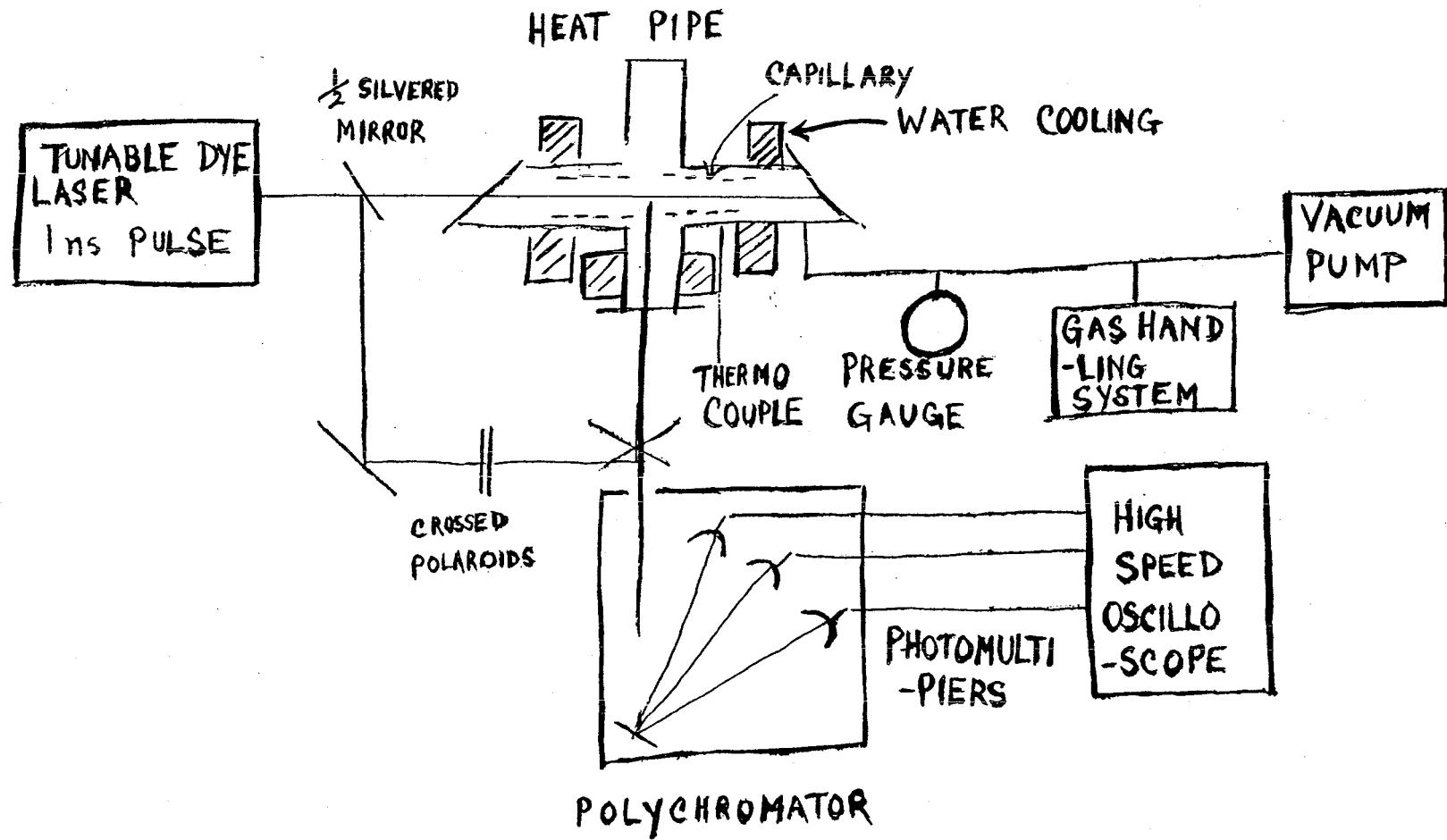


Figure 2.2 Method of measuring lifetimes

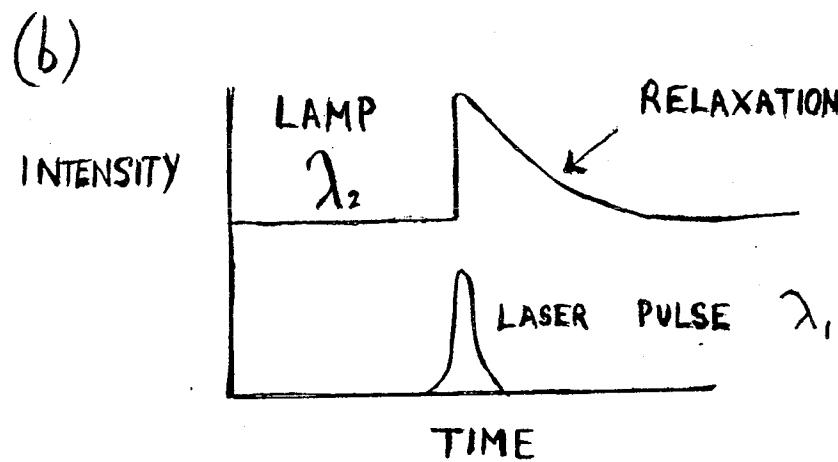
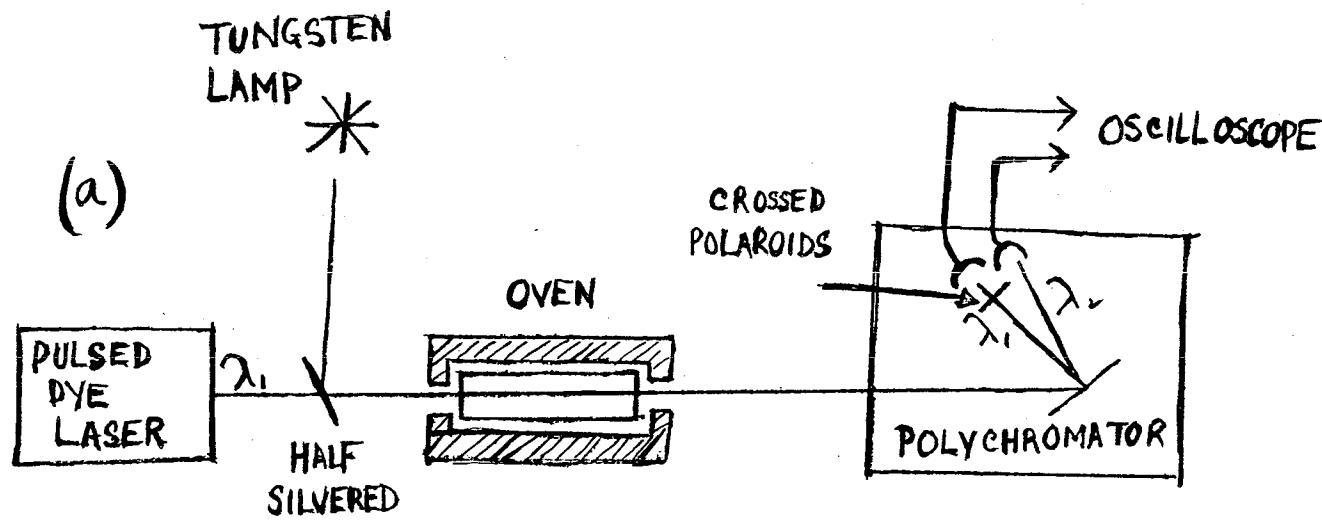


Figure 2.3 (a) Method for obtaining relaxation times for vibrational levels in the ground electronic state. Wavelength  $\lambda_2$  of figure 1 is monitored from the tungsten lamp. It corresponds to transition  $v'' \rightarrow v'$ . (b) The laser pulse at  $\lambda_1$  in figure 1 depletes level  $v'$  causing increased transmission of  $\lambda_2$ , which then decays as level  $v'$  is filled by collisions.

## THE POWER TO WEIGHT RATIO FOR SOLAR PUMPED LASERS

### SUMMARY

A general expression is obtained for the output power to weight ratio  $P_o/W$ , of solar pumped lasers assuming the weights of the collector and heat radiator are proportional to their areas. The ratio depends on the laser efficiency, and the temperature of the radiator. For IBr,  $C_3F_7I$  and  $Na_2$  lasers working at 600, 650, and 800 K respectively  $P_o/W$  is an order of magnitude higher for IBr and  $Na_2$  than for  $C_3F_7I$ .

### INTRODUCTION

One concept for the production of energy is to place solar collectors on orbiting space stations, and then transmit the energy via laser beams. Direct conversion by solar pumped gas lasers has been considered (1-3). A figure of merit, of importance for space missions, is the power to weight ratio of such laser systems.

The solar radiance is collected in a parabolic reflector and for a given output power, its area is determined by the laser efficiency. The power not converted into the laser beam must be dissipated by a heat radiator emitting by Stefan's Law; its area depends on its temperature. Here it is assumed a functional dependence exists between weight and area of both collector and radiator.

The overall efficiency of the laser can be decomposed into the product of several efficiencies which affect the collector and radiator in different ways, and the energy flow is discussed in the next section. The dependences of weight on area are then assumed which determines the weight for a given out-put power. Some comparisons will then be made of different laser systems.

### EFFICIENCY OF SOLAR PUMPED LASERS

The overall efficiency of solar pumped lasers can be subdivided into the product of four efficiencies. First if absorption occurs over a bandwidth  $\lambda_1$  to  $\lambda_2$ , the fraction of the solar spectrum used, or "solar utilization efficiency" is  $\eta_s$ :

$$\eta_S = \int_{\lambda_1}^{\lambda_2} \Phi(\lambda) d\lambda / \int_{\lambda_1}^{\infty} \Phi(\lambda) d\lambda$$

where  $\Phi(\lambda)$  is the solar radiance in watts per square meter between  $\lambda$  and  $\lambda + d\lambda$ .

The absorption efficiency  $\eta_A$  is that fraction of photons within the absorption bandwidth absorbed by the gas. At low pressure  $\eta_A = x\sigma_a(N)d$ , where  $x$  is the number of times the radiation passes through the gas by reflection,  $\sigma_a$  the absorption cross section,  $(N)$  the density of absorbers and  $d$  the thickness of gas. At high values of  $(N)d$  the absorption efficiency approaches 1.

Of the photons absorbed, only a fraction  $\eta_K$  end up in producing a lasing transition and  $\eta_K$  is the "kinetic efficiency." Finally the quantum efficiency  $\eta_Q$  is the ratio of the energy of the emitted photon to the average energy of the absorbed photons. The overall efficiency  $\eta$  is

$$\eta_S \eta_A \eta_K \eta_Q$$

The solar radiance is  $1.4 \text{ kwm}^{-2}$ , and if the output power is  $P_o$  (kW) then the area of the collector  $A_c$  is:

$$A_c = P_o / 1.4 \eta \text{ m}^2. \quad (1)$$

Of the input power  $P_i$ , then  $\eta_S \eta_A P_i$  is absorbed in the gas, and  $(1-\eta_S \eta_A) P_i$  is transmitted through. However of this unused portion, a fraction  $f$  is absorbed in the container walls and has to be emitted by the radiator. A fraction  $\eta_K$  of the power absorbed causes lasing, and the output power  $P_o$  is  $\eta_K P_i$ . The power that has to be radiated,  $P_r$  is:

$$P_r = P_o (\eta_S \eta_A + f) (1-\eta_K \eta_Q) / \eta \quad (2)$$

and if the radiator emits by Stefan's Law its area is:

$$A_r = P_r / \sigma \epsilon T^4 \quad (3)$$

where  $\sigma$  is Stefan's constant,  $\epsilon$  the emissivity and  $T$  the temperature of the emitting surface. It is assumed that efficient conduction by heat pipes causes  $T$  to approach the working temperature of the laser.

## OUTPUT POWER TO WEIGHT RATIO

Assumptions must now be made of the dependence of the weights of the collector and radiator on their respective areas. The collector is assumed to be either a spherical or cylindrical reflecting mirror, and its weight  $W_c$  is assumed proportional to its area:

$$W_c = \alpha A_c \quad (4)$$

Similarly, the radiator weight  $W_r$  is given by:

$$W_r = \beta A_r^n \quad (5)$$

The  $\alpha$  and  $\beta$  are constants of proportionality and  $n$  is a constant which is included to account for the weight of the heat pipes but probably does not differ greatly from 1.

From equations 1 through 5, the total weight  $W$  of the laser, collector and radiator is:

$$W = W_L + \frac{\alpha P_o}{1.4n} + \beta \frac{P_o (\eta_S \eta_A + f) (1 - \eta_K \eta_Q)^n}{\sigma \epsilon T^4} \quad (6)$$

where  $W_L$  is the weight of the laser. It is not unreasonable to assume  $W_L$  to be negligible with respect to the collector and radiator, and if so then the output power to weight ratio  $P_o/W$  is:

$$\frac{P_o}{W} = \frac{\eta_S \eta_A \eta_K \eta_Q}{\frac{\alpha}{1.4} + \beta \left[ \frac{\eta_S \eta_A + f}{\sigma \epsilon T^4} (1 - \eta_K \eta_Q) \right]^n} \quad ; \quad W_L \ll W \quad (7)$$

The ratio is increased if  $\alpha, \beta \rightarrow 0$  and  $T$  is as high as possible. The dependence on the efficiencies is  $P_o/W(x, y)$  where  $x = \eta_S \eta_A$  and  $y = \eta_K \eta_Q$ . A plot of  $P_o/W$  on an  $xy$  plane is shown in figure 3.1, for  $f = 0$ ,  $n = 1$ ,  $\alpha = 1$  and  $\beta = 1$  and 10, the latter being an extreme case for illustrative purposes. The range of  $x$  and  $y$  is only 0-0.3 corresponding to actual efficiencies in practice. The temperatures chosen are from 300 to 1500 K in steps of 300.

For both cases  $P_o/W$  is either constant or increases monotonically with both  $x$  and  $y$ , and the highest  $P_o/W$  occurs in the right hand corner of

the diagrams. The effect of  $\beta/\alpha$  is large at low temperatures, (comparing Figure 3.1a and 3.1b) and here  $P_o/W \approx \alpha/\beta$ . At high temperatures the  $T^4$  dependence quickly makes the area required for the radiator small and  $P_o/W$  is independent of both  $T$  and  $\beta$ .

Assuming that  $\alpha$  and  $\beta$  which are determined by structure design are constants for all lasers, then equation 7 enables a comparison to be made for different lasing materials provided the various efficiencies are known. A comparison of IBr,  $C_3F_7I$  and  $Na_2$  lasers is shown in table 1. It is assumed that the gas pressure and depth are sufficiently high that  $\eta_A = 1$ . The kinetic efficiency of  $C_3F_7I$  is not known and is assumed as 1. That of  $Na_2$  is also doubtful. The value of  $\alpha P_o/W$  is calculated from equation 7 assuming  $\alpha = \beta$ ,  $f = 0$ . The temperatures are approximate working temperatures.

#### CONCLUSIONS

Assuming that a solar pumped laser has a radiation collector and heat emitter whose weights are proportional to their areas, and that the weight of the actual laser is negligible in comparison, a relationship is obtained for the output power per unit weight. Table 1 shows that the materials IBr, and Na have a power to weight ratio an order of magnitude higher than  $C_3F_7I$ . In these calculations the fraction  $f$  absorbed by the container walls was made zero. The value of  $f$  is of importance when the working temperatures are low. If it were 0.05 for example, then it has to be added to  $\eta_S \eta_A$ , and make things still worse for  $C_3F_7I$  which has the lowest  $\eta_S \eta_A$ .

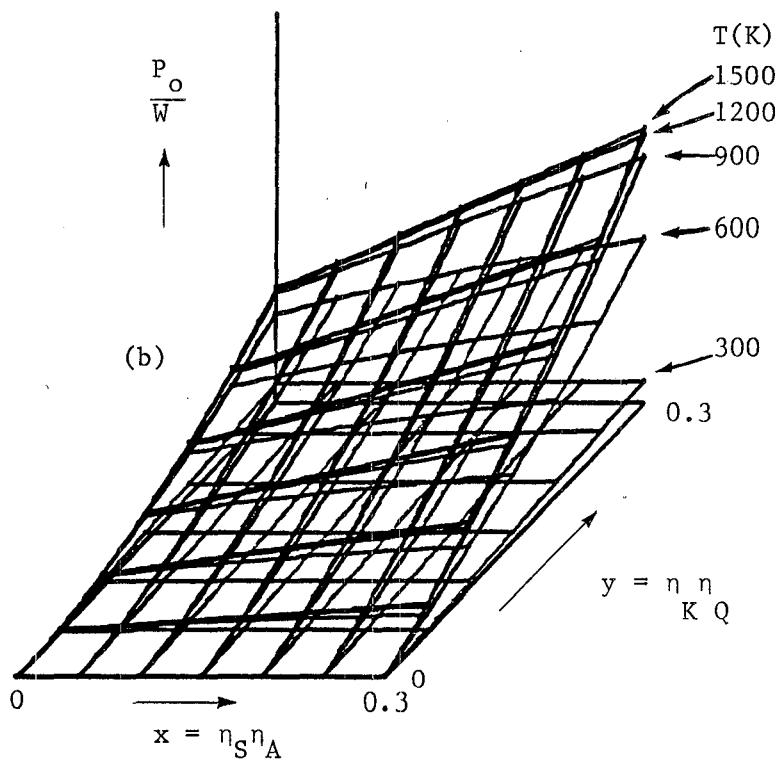
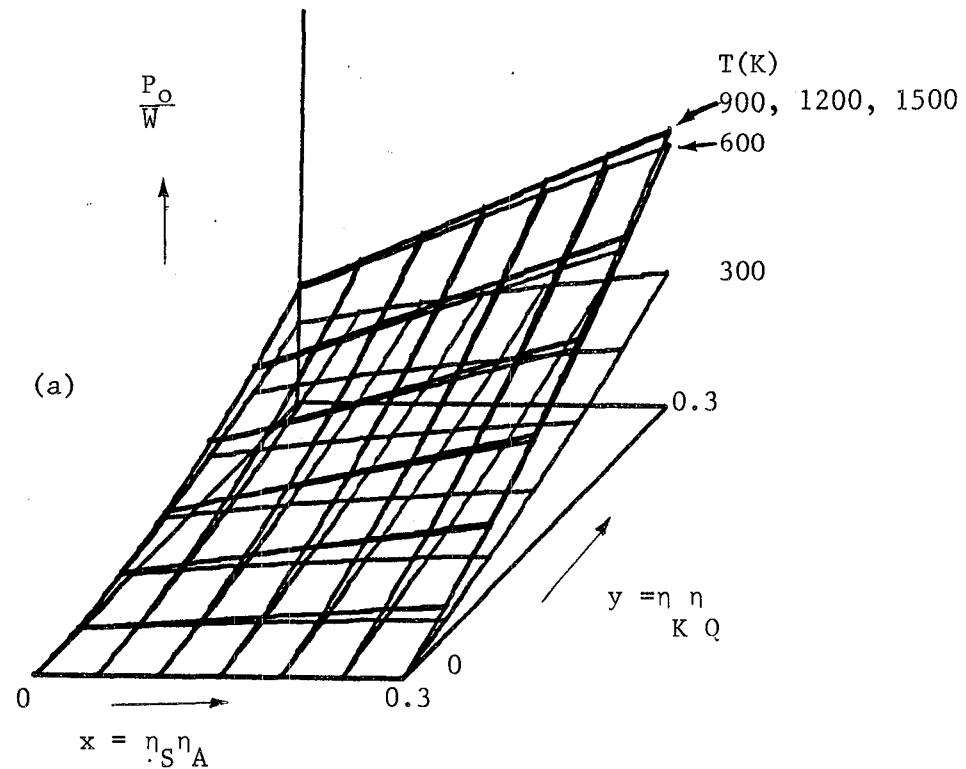


Figure 3.1 Plot of  $P_o/W$  on an  $x, y$  plane for different temperatures.  
 $n = 1, \alpha = 1$ , (a)  $\beta = 1$ , (b)  $\beta = 10$  an extreme case.

Table 1

## Comparison of Various Lasing Materials in Solar Pumped Lasers

Material	$\eta_S$	$\eta_A$	$\eta_K$	$\eta_Q$	$\eta$	T(K)	$\alpha P_o/W$
IBr	0.12	1	0.57	0.18	$1.2 \times 10^{-2}$	600	$1.75 \times 10^{-2}$
$C_3 F_7 I$	$4.1 \times 10^{-3}$	1	? 1	0.22	$1 \times 10^{-3}$	650	$1.3 \times 10^{-3}$
$Na_2$	$2 \times 10^{-2}$	1	? 0.5	? 1	$1 \times 10^{-2}$	800	$1.4 \times 10^{-2}$

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